

Temperature dependence of optical linewidth in single InAs quantum dots

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We consider the temperature dependence of the exciton linewidth in single InAs self-assembled quantum dots. We show that in cases where etched mesas are used to isolate the dots, the magnitude of the linear temperature coefficient and its dependence on mesa size are described well by exciton scattering by acoustic phonons whose lifetimes are given by phonon scattering from the mesa interfaces. This work shows that phonon scattering at interfaces and surfaces typically present in quantum dot structures can be important in dynamical processes in quantum dot systems.

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The interaction of excitons with phonons in semiconductor quantum dots determines the temperature dependence of the optical response and contributes to the dephasing of the optically excited quantum states of the dots. These optical properties are of current interest in connection with proposed use of quantum dots in implementations of quantum computing. At low temperatures the photoluminescence spectra of single dots have a sharp central feature and a broad background.¹⁻⁵ The problem of phonon effects in the optical spectra associated with bound electronic states in a crystal was first treated in the context of spectra of impurities⁶⁻⁹ and bound excitons in bulk semiconductors.⁹ In terms of a perturbation treatment of the electron-phonon interaction in the case of acoustic phonons the central peak in the absorption or emission spectrum is associated with the term involving no phonons and commonly referred to as the zero-phonon line (ZPL). It is an optical analog of the Mössbauer line in the impurity nuclei transitions.⁸

The experimentally observed peak has a width that was found to increase linearly with temperature at $T < 50$ K in many single dot studies.¹⁻⁴ The inverse of the ZPL width is related to the rate of the polarization decay, i.e., the dephasing time, and has been studied for single dots and for arrays of dots in four-wave mixing experiments.^{2,10} In recent four-wave mixing experiments on arrays of dots the temperature dependence of the linewidth was found to be of an exponential activated form rather than linear.^{11,12} Two different mechanisms involving off-diagonal transitions between quantum dot electronic states have been proposed as explanations of ZPL broadening, real transitions from ground to excited states in larger dots¹³ such as those resulting from interface fluctuations during molecular beam epitaxy (MBE) growth and virtual transitions giving higher order terms in exciton-phonon interaction.¹⁴ Using realistic values of interaction parameters, we find that for smaller dots, such as those resulting from strain controlled self-organized growth at interfaces, neither of these mechanisms explains the order of magnitude of the temperature dependence of the optical linewidth observed in single dots.

In this work we consider the case of single self-assembled dots, and in particular dots in etched mesas introduced to isolate the single dots.^{1,5} In the more recent of these studies the luminescence ZPL width was obtained as a function of temperature for various values of the linear size of the mesa.

The temperature dependence was found to be fitted well by a straight line $\Gamma(T) = \Gamma(0) + \alpha T$ with slope α monotonically decreasing with increasing mesa size. In this work we define the linewidth as a full width at half maximum. The observed increase of the low temperature linewidth $\Gamma(0)$ with decreasing mesa size could have a contribution from fluctuating charge occupation at the lateral mesa interfaces. However, because carriers occupy deep traps at the side walls with large activation energy, they would contribute to weak exponentially activated variation rather than a linear T dependence of the linewidth. Therefore the observed mesa size dependence of α does not originate in fluctuations of interface charges.⁵

We use the independent boson model in order to evaluate the optical spectra of localized electrons coupled to acoustic phonons.^{8,9} In the subspace of the electron-hole pairs the interaction can be written in terms of phonon and confined exciton operators a and B . To the linear order in phonon operators

$$H_{\text{int}} = \sum_{n,m} \hat{B}_n^\dagger \hat{B}_m \sum_{\mathbf{q},\sigma} M_{nm}(\mathbf{q},\sigma) (\hat{a}_{\mathbf{q},\sigma} + \hat{a}_{-\mathbf{q},\sigma}^\dagger), \quad (1)$$

where M_{nm} is the interaction matrix element. The summations are over the confined states of the interacting electron-hole pair in the quantum dot and the wave vector and polarization of bulk phonons. If only the diagonal part of H_{int} , which we shall call H_{int}^d , is retained, one obtains an exactly solvable model. This model was used in Ref. 9 to evaluate the spectral function of a bound exciton in a bulk semiconductor. We shall treat the off-diagonal part as a perturbation. Following Ref. 8, we shall also take into account the finite lifetime of the phonons and explicitly consider various phonon lifetime limiting processes.

The absorption spectrum can be obtained as a Fourier transformation of a one-particle time-dependent exciton Green's function. This is equivalent to using a two-particle interband correlation function in the Kubo formula for linear response in terms of electron operators¹⁵ and gives time dependence of light induced polarization. Using the method of linked cluster expansion,¹⁵ one can easily generalize the treatment in Ref. 9 to include finite phonon lifetimes in the derivation of the electronic correlation functions. Assuming that phonons are in thermal equilibrium, the phonon correlation functions are

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$$\langle \hat{a}_q^\dagger(t) \hat{a}_q(0) \rangle = \exp(i\omega_q t - \gamma_q t) n_q, \quad (2)$$

$$\langle \hat{a}_q(0) \hat{a}_q^\dagger(t) \rangle = \exp(i\omega_q t - \gamma_q t) (n_q + 1), \quad t \geq 0,$$

where $n_q = [\exp(\hbar\omega_q/k_B T) - 1]^{-1}$ and the phonon lifetime $\tau_q = \hbar/\gamma_q$. We then obtain the following result for the ground state exciton Green's function.

$$G(\omega) = -i \int_0^\infty dt e^{i(\omega - \omega_0 + i\varepsilon)t} e^{f(t)} (\varepsilon \rightarrow +0), \quad (3)$$

where $\hbar\omega_0$ is the energy of the exciton state

$$f(t) = (i\Delta - \Gamma/2)t/\hbar - \tilde{f}(t),$$

$$\Delta = \sum_{\vec{q}, \sigma} \frac{|M_{00}(\vec{q}, \sigma)|^2 \omega_{\vec{q}\sigma}}{\hbar^2 \omega_{\vec{q}\sigma}^2 + \gamma_{\vec{q}}^2}, \quad (4)$$

$$\Gamma/2 = \sum_{\vec{q}, \sigma} \frac{|M_{00}(\vec{q}, \sigma)|^2 \gamma_{\vec{q}}}{\hbar^2 \omega_{\vec{q}\sigma}^2 + \gamma_{\vec{q}}^2} (2n_q + 1), \quad (5)$$

$$\tilde{f}(t) = \sum_{\vec{q}, \sigma} \frac{|M_{00}(\vec{q}, \sigma)|^2 (\hbar^2 \omega_{\vec{q}\sigma}^2 + 2i\hbar \gamma_{\vec{q}} \omega_{\vec{q}\sigma})}{(\hbar^2 \omega_{\vec{q}\sigma}^2 + \gamma_{\vec{q}}^2)^2}$$

$$\times ((2n_q + 1)(1 - e^{-\gamma_q t/\hbar}) + \{[1 - \cos(\omega_{\vec{q}\sigma} t)](2n_q + 1)$$

$$- i \sin(\omega_{\vec{q}\sigma} t)\} e^{-\gamma_q t/\hbar}), \quad (6)$$

and the temperature dependence enters through $2n_q + 1 = \coth(\hbar\omega_q/k_B T)$.

In the evaluation of the exciton-phonon interaction matrix elements we consider a cylindrically shaped $\text{In}_x\text{Ga}_{1-x}\text{As}$ quantum dot (a disk) of height L and base radius R . The Coulomb correlation in the electron-hole pair is included through variational wave functions for the ground and excited pair states. We construct exciton states out of one-particle confined states, which are eigenfunctions of z projection of orbital angular momentum, with z chosen along the growth direction. The one particle confinement potentials obtained from the band gap discontinuities U_i , $i=e, h$ can be written as $V_i(r_i, z_i) = U_i \theta(r_i - R) + U_i \theta(|z_i| - L/2) + \delta V_i(r_i, z_i)$ where the nonseparable part $\delta V_i(r, z) = U_i \theta(|z| - L/2) \theta(r_i - R)$ can be treated as a perturbation.

For the disklike case of $2R > L$ we neglect the lateral tunneling. The anisotropy of the valence band should be taken into account: $m_{xy} \neq m_z$. For the ground state of the electron-hole pair we take $\Psi_0(\mathbf{r}_e, \mathbf{r}_h, z_e, z_h) = A_0 f_1^e(z_e) f_1^h(z_h) J_0(r_e x_{01}/R) J_0(r_h x_{01}/R) \exp(-\alpha_0 |\mathbf{r}_e - \mathbf{r}_h|)$, where functions $f_n(z)$ refer to vertical confinement, $J_m(r)$ are Bessel functions. For typical cases where the hole in-plane mass m_{xy} is substantially larger than the electron mass, we take the following variational form for the degenerate first excited state $\Psi_{\pm 1}(\mathbf{r}_e, \mathbf{r}_h, z_e, z_h) = A_1 f_1^e(z_e) f_1^h(z_h) J_0(r_e x_{01}/R) J_1(r_h x_{11}/R) \exp(-\alpha_1 |\mathbf{r}_e - \mathbf{r}_h|) \times \exp(\pm i\varphi_h)$, where φ_h is the in-plane angular coordinate of the hole. In these two expressions α_0 and α_1 are variational parameters, x_{mk} are positive zeros of Bessel function $J_m(x)$.

In the case of an $\text{In}_{0.6}\text{Ga}_{0.4}\text{As}/\text{GaAs}$ dot the band gap discontinuity $\Delta E_g \approx 0.74$ eV and we took 40%–60% split between valence and conduction band edges in evaluation of confining potentials U_e and U_h . The electron mass in the dot

$m_e = 0.0404$ in units of free-electron mass m_0 , and 0.0665 in the barrier material. The hole mass parameters in the dot are $m_{hz} = 0.34$, $m_{hxy} = 0.09$. The value of the heavy hole mass for the in-plane dispersion was obtained from the analytical solution for the infinite barrier case with the appropriate values of Luttinger parameters.¹⁶ For a disk with dimensions $L = 30$ Å and $R = 100$ Å we obtain $\alpha_0 R = 0.15$, $\alpha_1 R = 0.14$. The average distance between the electron and the hole in the ground state is $\langle |\mathbf{r}_e - \mathbf{r}_h|^2 \rangle^{1/2} = 75$ Å. The excited state and the ground state are separated by 37.7 meV. The effect of the Coulomb interaction is to shift the ground and excited state energies by similar amounts, −12.6 meV and −11 meV, respectively. When the parameters are such that $1/\alpha_{0,1} \gg R$, the Coulomb correlation of the electron and hole is small compared to their confinement by the band discontinuities. In such a case one can use one-particle wave functions in the estimate of interaction matrix elements.

At lower temperatures we consider the interaction of electrons and holes with acoustic phonons only. We included the band deformation potential interaction,¹⁷ the piezoelectric interaction, and the electron-phonon interaction produced by the phonon induced deformation of the confining potential, also known as the “ripple mechanism.”¹⁸ A formal expansion of $\exp[f(t)]$ in Eq. (3) produces a spectrum in the form of the sum of terms that involve n phonon transitions with $n = 0, 1, 2, \dots$. At low temperature the spectrum has a form of ZPL plus a broad background from many-phonon transitions. The overall line shape can be deduced by considering the short and long time behavior of the integrand in Eq. (3). If we assumed an infinite phonon lifetime, i.e., set $\gamma = 0$ in Eq. (2), then $\Gamma = 0$ and at short times $t \text{ Re } \tilde{f}(t) \propto -t^2$, while at large $t \text{ Re } \tilde{f}(t) \propto 1/t$. In such a case the zero-phonon line is not broadened by the diagonal part of the exciton-phonon interactions.

Before evaluating the effect of the finite phonon lifetime in Eq. (3) we consider the effects of the off diagonal terms in H_{int} in Eq. (1). The real transitions to excited states can contribute to exciton dephasing in larger disklike dots.¹³ The order of magnitude of the corresponding contribution to the exciton linewidth, $\Gamma(0 \rightarrow 1)$, can be obtained from the ground to excited confined exciton state with one-phonon absorption. For the $R = 10$ nm dot we obtained that at $T = 100$ K $\Gamma(0 \rightarrow 1) \sim O(10^{-4} \text{ } \mu\text{eV})$. Recently the effect of virtual transitions in H_{int} was treated by deriving an effective quadratic coupling in exciton-phonon interaction.¹⁴ Using the realistic values for deformation potentials, we estimated the corresponding contributions from Ref. 14 to be an order of magnitude smaller than the observed temperature dependent part of ZPL width in single dot experiments.⁵ We conclude then that the contribution of the off diagonal terms in Eq. (1) to the experimentally observed linewidth for smaller dots is negligible. It was suggested in Ref. 8 that direct anharmonic terms should be included in Eq. (1). It was later shown, however, that the inclusion of terms quadratic in phonon operators in the diagonal part of the exciton-phonon interaction does not lead to the broadening of the zero-phonon line in case of infinite phonon lifetime.¹⁹

When the phonon linewidth γ is included in the treatment, the resulting width Γ of the zero-phonon peak in the absorp-

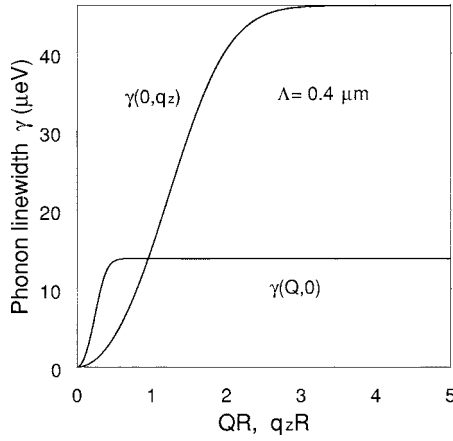


FIG. 1. Phonon linewidth induced by the boundary scattering, shown as a function of wave vector for lateral and growth directions.

tion spectrum of the dot is given in Eq. (5). We evaluated several sources of the finite lifetime of acoustic phonons: anharmonicity induced three-phonon processes,²⁰ scattering of phonons by impurities and alloy fluctuations, and boundary phonon scattering.²¹ In cases where etched mesas are present, we find that well below room temperature, the acoustic phonon lifetimes are limited primarily by scattering at the side surfaces of the mesa for smaller mesas and at the top surface of the structure for large mesas. We also find that in the evaluation of Eq. (5) with γ_q given by the boundary phonon scattering, almost all of the resulting value of zero-phonon peak linewidth comes from the band deformation potentials which we took here to be -9.3 eV for the conduction band and 3 eV for the valence band.¹⁷

We obtain the phonon lifetime from the steady state Boltzmann equation for phonons. This is similar to the evaluation of boundary scattering in the thermal conductivity.²² Results similar to those given here are obtained by evaluating the eigenmodes of an acoustic waveguide with rough surfaces. The approach commonly used in the theory of wave scattering from rough surfaces is that these surfaces are ergodic.²³ In the case of sound scattering this is justified if the phonon spectrum is continuous.²⁴ Then the eigenfrequencies acquire an imaginary part, even when the scattering is completely elastic.^{25–27} Using the relaxation time approximation, we obtained in the case of completely diffusive surfaces $1/\tau_q^0 \approx 2v_\perp/\Lambda$ for scattering from the mesa sides and $1/\tau_q^0 \approx v_z/\Lambda_z$ for scattering from the top surface of the GaAs top layer. Here v_\perp and v_z are the in plane and z components of the phonon velocity, Λ is the side length of the mesa which was assumed to have a square cross section, and Λ_z is the thickness of the top layer. For example, a completely diffusive scattering of phonons by the faces of $0.4 \mu\text{m}$ mesa gives $\gamma^0 \sim 10 \mu\text{eV}$. To compare this to the three-phonon processes, we can consider the most efficient of the latter, the Landau-Rumer process of relaxation of a transverse acoustic mode into thermal longitudinal modes²⁰ giving $\hbar/\tau_q = 6g^2\omega_q(k_B T)^4(1+2v_T/3v_L)/(\pi\rho v_T v_L^4 \hbar^2)$, which we evaluate at $q=0.2 \text{ nm}^{-1}$ and take Grüneisen constant $g=2$. This gives about $0.5 \mu\text{eV}$ at 50 K and becomes comparable to the boundary scattering phonon linewidth only above 100 K .

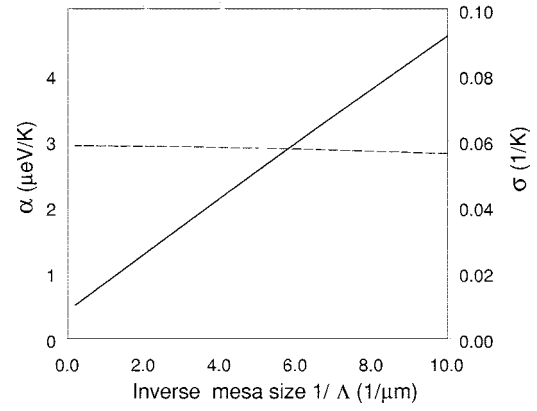


FIG. 2. Coefficients of the linear temperature dependence of the ZPL width (α , solid line) and of Huang-Rhys factor (σ , broken line), shown as functions of the inverse mesa lateral size.

Thus the anharmonic phonon-phonon interactions are not important in the evaluation of Eq. (3) below 100 K .

In the case of partially diffusive surfaces we use a specularly parameter defined by the boundary condition for the nonequilibrium part $\delta n(\vec{q}, \vec{r})$ of the phonon distribution function at a particular boundary, e.g., $\delta n(q_z, \Lambda_z) = p(\vec{q})\delta n(-q_z, \Lambda_z)$ for the scattering from the top horizontal surface at $z = \Lambda_z$. As was shown in Ref. 21, $\gamma_q = \gamma_q^0[1 - p(\vec{q})]/[1 + p(\vec{q})]$ and one can use a Gaussian approximation for $p(q)$: $p(q) = \exp(-4\pi\delta^2 q^2)$ where the asperity parameter δ is the average height of the surface bumps.²¹

In the evaluation of Eq. (5) we assume the following linear dimensions: $R=10 \text{ nm}$, $L=6 \text{ nm}$ for the dot, $\Lambda_z=6 \text{ nm}$, asperity parameters $\delta=12 \text{ nm}$ for the mesa sides and $\delta_z=1 \text{ nm}$ for the top (epitaxial) surface. The phonon linewidth is shown in Fig. 1 as a function of wave vector $\vec{q}=(\vec{Q}, q_z)$ in two directions in the momentum space for $\Lambda=0.4 \mu\text{m}$.

The width of ZPL, Γ , as a function of temperature T and mesa size Λ was obtained from Eq. (5). We find that in evaluation of electron-phonon matrix elements in Eq. (5) almost all off the value of the width of the zero phonon line comes from the deformation potential interaction, while the ripple mechanism contributes about 1% and the piezoelectric interaction contributes much less than that. The height of

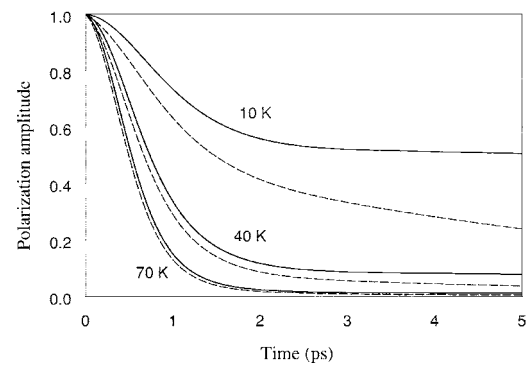


FIG. 3. Optically induced polarization amplitude for a single quantum dot in $0.4 \mu\text{m}$ mesa, normalized to one for three values of temperature and two values of zero temperature linewidth, $\Gamma(0) = 1 \mu\text{eV}$ (solid lines) and $\Gamma(0) = 100 \mu\text{eV}$ (broken lines).

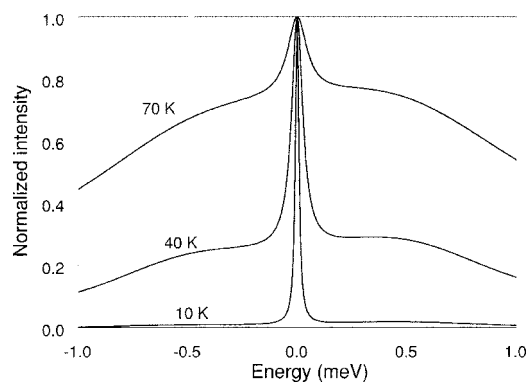


FIG. 4. Optical absorption for a single quantum dot in $0.4 \mu\text{m}$ mesa, normalized to one for three values of temperature and centered at the ZPL energy, with $\Gamma(0)=1 \mu\text{eV}$.

ZPL is determined by a Debye-Waller factor e^{-S} with Huang-Rhys factor^{7,9} S defined as the real part of the time independent term in Eq. (6)

$$S = \sum_{\vec{q}, \sigma} \frac{|M_{00}(\vec{q}, \sigma)|^2 \hbar^2 \omega_{\vec{q}\sigma}^2}{(\hbar^2 \omega_{\vec{q}\sigma}^2 + \gamma_{\vec{q}}^2)^2} (2n_{\vec{q}} + 1). \quad (7)$$

At temperatures above 5 K we find that both the linewidth and Huang-Rhys factor are given by nearly linear functions of T , $\Gamma(T) - \Gamma(0) = \alpha T$ and $S(T) - S(0) = \sigma T$, and we evaluated the linear coefficients α and σ as functions of the mesa size Λ . The linewidth coefficient α varies approximately linearly with the inverse of the mesa size while the Huang-Rhys coefficient σ varies only slightly with the mesa size. The coefficients are shown in Fig. 2 as functions of $1/\Lambda$.

In the evaluation of the total linewidth we should also include the sample dependent term $\Gamma(0)$ which has been found to vary from $1 \mu\text{eV}$ to $200 \mu\text{eV}$ for single dots in

mesas.^{1,5} The time dependence of optically induced polarization amplitude²⁸ is given by $\exp\{\text{Re}[f(t)]\}$ and is shown in Fig. 3 in normalized form for three values of temperature, the mesa size $\Lambda=0.4 \mu\text{m}$, and for two values of $\Gamma(0)$. The optical absorption is given by $\text{Im} G(\omega)$ and is shown in Fig. 4 normalized to one for the same values of T and $\Gamma(0)$ as in Fig. 3. The small polariton shift Δ is of order of temperature dependent part of the linewidth. The emission spectra can be obtained easily by assuming an equilibrium distribution of photons in the emitted light.¹⁵ The spectra are similar to those in Fig. 4 for lower temperatures except for the appearance of a small asymmetry.

Comparing our results with the experiments on single dots in mesas we conclude that a linear temperature dependence of the ZPL results mostly from deformation potential interaction of confined exciton with acoustic phonons that are scattered by the interfaces, with scattering from the surfaces formed in lateral fabrication giving the dominant contribution to the broadening of the acoustic phonons.

We also point out that even for those quantum dot experiments that do not have explicit etched mesas, the effects of interface induced phonon linewidths discussed here contribute to the ZPL broadening of the exciton due to phonon scattering at interfaces and surfaces formed during growth and fabrication. In such cases one obtains a smaller value for the coefficient α than those obtained here, which is consistent with the corresponding experiments.² In addition, the scattering of phonons at interfaces, which are typically present in quantum dot structures, can affect a wide range of phonon induced dynamical processes in quantum dot systems.

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